



Design of a new phase boundary to enhance the piezoelectric properties of lead-free $(\text{K}_{0.48}\text{Na}_{0.48}\text{Li}_{0.04})(\text{Nb}_{0.95}\text{Sb}_{0.05})\text{O}_3 - \text{Bi}_{0.5}\text{K}_{0.5}\text{HfO}_3$ ternary ceramics

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Abstract

In this study, a new phase boundary was successfully established in $(1-x)(\text{K}_{0.48}\text{Na}_{0.48}\text{Li}_{0.04})(\text{Nb}_{0.95}\text{Sb}_{0.05})\text{O}_3 - x\text{Bi}_{0.5}\text{K}_{0.5}\text{HfO}_3$ (KNLNS- x BKH) ternary ceramics. Changes in phase structure were verified by the X-ray diffraction (XRD) analysis and Raman scattering measurements of KNLNS- x BKH with varying x contents. The results indicate that KNLNS- x BKH exhibits an orthorhombic phase ($Amm2$) at $x=0$, a mixed orthorhombic-tetragonal phase ($Amm2 + P4mm$) at $x=0.01-0.02$, a mixed rhombohedral-tetragonal ($R-T$) phase ($R3m + P4mm$) at $x=0.03-0.04$, and ultimately to a single rhombohedral phase ($R3m$) at $x=0.05$. Under optimal conditions, the $(1-x)$ KNLNS- x BKH ceramics demonstrated remarkable electrical properties, including a density of 4.58 g/cm^3 , a piezoelectric constant (d_{33}) of 238 pC/N , a remanent polarization of (P_r) of $15.4 \text{ } \mu\text{C/cm}^2$ and a high Curie temperature (T_C) of nearly $310 \text{ }^\circ\text{C}$ at $x=0.03$. These properties are attributed to the establishment of a new rhombohedral (R)-tetragonal (T) phase boundary with microstructures characterized by high density, large grain size, and low porosity. Furthermore, KNLNS- x BKH ceramics with $x=0.05$ exhibited a high recoverable energy density ($W_{\text{rec}} = 0.26 \text{ J/cm}^3$) and a large energy storage efficiency ($\eta=56.8\%$) under a low electric field of 20 kV/cm , representing a significant advancement towards the development of practical applications.

Keywords KNLNS-BKH · New phase boundary · Lead-free ceramics · Ternary ceramics

1 Introduction

The $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$ (PZT)-based materials exhibit outstanding piezoelectric properties at the morphological phase boundary (MPB), which have been extensively demonstrated in a wide range of applications such as sensors, transducers, electronic devices, actuators, high power ultrasound, and

piezoelectric transformers [1–3]. Consequently, PZT-based materials have dominated the global piezoelectric materials market for several decades. Nevertheless, the high lead content, exceeding 60 wt% in PZT-based materials, poses serious risks to the environment and human health. This concern has driven increasing research efforts toward the development of lead-free ceramics, such as BaTiO_3 , $(\text{Bi}, \text{Na})\text{TiO}_3$, and $(\text{K}, \text{Na})\text{NbO}_3$ based systems [4, 5].

Among them, $(\text{K}, \text{Na})\text{NbO}_3$ (KNN) based ceramics, composed of ferroelectric KNbO_3 and antiferroelectric NaNbO_3 , have garnered significant interest from several materials scientists. Notably, these ceramics are considered one of the most promising candidates to replace PZT-based ceramics, which are harmful to the environment and human health [6–8]. Additionally, the electrical properties of KNN ceramics can be substantially improved, achieving high piezoelectric performance upon the establishment of a new phase boundary. Specifically, the design of a new phase boundary has emerged as the most extensively studied theme in lead-free

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(K, Na)NbO₃-based ceramics, which is widely regarded as an effective strategy for performance modification [4, 9].

A novel strategy has been established to form a new rhombohedral (*R*)-tetragonal (*T*) phase boundary in (K, Na)NbO₃ ceramics. This approach is generally regarded as including polymorphic phase boundary (PPT) through the process of single doping (such as Sb⁵⁺, Ta⁵⁺, Li⁺, Mn³⁺) in KNN ceramics, which is analogous to the morphological phase boundary (MPB) observed in PZT ceramics. Moreover, the co-doping of Li⁺ and Sb⁵⁺ ions into (K, Na)NbO₃ ceramics leads has been demonstrated to shift the PPT to lower temperature compared with pristine KNN ceramics, thereby significantly enhancing the piezoelectric properties, in agreement with the results reported by Tu et al. [10, 11]. Notably, adding an ABO₃-type component (such as Bi_{0.5}(Na_xK_{1-x})ZrO₃, (Bi_{0.5}K_{0.5})HfO₃, (Bi_{0.5}Na_{0.5})HfO₃, CaZrO₃, BaZrO₃, and BaTiO₃) to the KNN-based ceramics tune the rhombohedral-orthorhombic transition temperature (*T*_{R-O}) and orthorhombic-tetragonal transition temperature (*T*_{O-T}) to room temperature simultaneously, thereby improving their piezoelectric properties [12, 13]. It has been reported that various chemical ABO₃-type substitutions into (K, Na)NbO₃ or (K, Na)(Nb, Sb)O₃ can decrease the *O-T* phase transition temperature (*T*_{O-T}), enhance the *R-O* phase transition temperature (*T*_{R-O}), and thus limit the temperature range of the orthorhombic phase [14, 15]. According to Zhang et al. [14], 0.955(K_{0.48}Na_{0.52})(Nb_{0.96}Sb_{0.04})O₃-0.045(Bi_{0.50}Na_{0.50})HfO₃ ceramics exhibited excellent piezoelectric properties (*d*₃₃=540 pC/N) owing to the coexistence of the rhombohedral-orthorhombic-tetragonal (*R-O-T*) phase at room temperature. As reported by Tao et al. [16], 0.965K_{0.45}Na_{0.55}Nb_{0.96}Sb_{0.04}O₃-0.035Bi_{0.5}Na_{0.5}HfO₃ ceramics at the *R-T* phase boundary exhibited a large *d*₃₃ value of 418 pC/N, a high *k*_p of 0.45, and a *T*_C of 242 °C. Furthermore, Chen et al. [17] reported that (1-*x*)(K_{0.48}Na_{0.48}Li_{0.04})(Nb_{0.95}Sb_{0.05})O₃-*x*(Bi_{0.5}Na_{0.5})HfO₃ ceramics with compositions near the orthorhombic-tetragonal phase boundary (*x*=0.005) exhibited maximum values of the piezoelectric constant *d*₃₃ and planar and thickness electromechanical coupling coefficients (*k*_p and *k*_t) of 246 pC/N, 0.435, and 0.554, respectively.

However, to our knowledge, the Bi_{0.5}K_{0.5}HfO₃ (BKH) component has been used less often than (Bi_{0.50}Na_{0.50})HfO₃ for establishing phase boundaries for KNN-based ceramics. This is attributed to the highly hygroscopic nature of the element K. Consequently, the enhancement of K in the KNN-based ceramics is often less emphasized than that of Na. On the other hand, recent studies have demonstrated that the ratio of K is less than Na in the KNN-based ceramics [10, 11]. Interestingly, Liu et al. [18] have confirmed that Bi_{0.5}K_{0.5}HfO₃ is beneficial and can significantly improve

the piezoelectric properties of KNN-based ceramics through appropriate composition design. Furthermore, the co-doping of Bi³⁺, K⁺, and Hf⁴⁺ ions in KNN-based ceramics is expected to play a crucial role in improving the sintering temperature, microstructure, and energy storage performance of (K, Na)(Nb, Sb)O₃ ceramics, which aligns with the findings of Tao et al. [16]. This study presents a new phase boundary in (1-*x*)KNLNS-*x*BKH ternary ceramics. This work aims to comprehensively investigate the effect of BKH content on the physical properties of BKH-doped (K_{0.48}Na_{0.48}Li_{0.04})(Nb_{0.95}Sb_{0.05})O₃ ceramics, determine the optimal BKH content, and explain the formation of new phase boundaries to improve the electrical properties of KNLNS-based materials.

2 Experimental

In this study, the (1-*x*)(K_{0.48}Na_{0.48}Li_{0.04})(Nb_{0.95}Sb_{0.05})O₃-*x*Bi_{0.5}K_{0.5}HfO₃ (KNLNS-*x*BKH, *x*=0, 0.01, 0.02, 0.03, 0.04, and 0.05) ceramics were fabricated using a two-step sintering method. The starting materials in this stage include Na₂CO₃, K₂CO₃, Li₂CO₃, Nb₂O₅, Sb₂O₃, Bi₂O₃, and HfO₂, with purity ≥ 99.5%. This mixture was milled together for 20 h using a planetary ball mill in ethanol. Subsequently, the mixture was dried and calcined at 850 °C for 2 h to form the compound KNLNS-BKH. The mixed KNLNS-BKH powders were ball-milled again for 20 h, dried, and pressed into disks with a thickness of 1.5 mm and a diameter of 12 mm under 1 T/cm² to form green samples. The two-step sintering process of KNLNS-BKH samples was conducted as follows. Starting from room temperature (*RT*), the samples were heated to 1150 °C at a heating rate of 5 °C/min and maintained at 1150 °C for 5 min. Subsequently, the temperature was reduced to a lower temperature at *T*₂=1050 °C at a heating rate of 20 °C/min and maintained for 4 h to form BKH-doped KNLNS ceramics. Finally, the samples were cooled to *RT* at a heating rate of 5 °C/min, thereby completing the ceramic sintering process.

The crystal structures and surface morphologies of the samples were examined using X-ray diffraction (XRD) and scanning electron microscopy (SEM), respectively. The density of samples was measured using the Archimedes' method. The ferroelectric properties of ceramics were examined using the Sawyer-Tower method. The capacitance and dielectric loss of the KNLNS-BKH samples were measured using an impedance analyzer (RLC HIOKI 3532) over a temperature range of 25–400 °C. Finally, the samples were poled in a silicone oil bath at various temperatures (25, 35, 45, 55, and 65 °C) under applied electric field of 25, 30, and 35 kV/cm for 25 min to measure their piezoelectric properties.

3 Results and discussion

Figure 1(a) presents the major findings of the XRD analysis of KNLNS–BKH ceramics within the 2θ range of $20\text{--}70^\circ$. The as-synthesized KNLNS–BKH ceramics mainly exhibited a perovskite structure, and no other impurities were observed at lower x contents ($0 < x \leq 0.04$). Nevertheless, secondary phase peaks are observed when $x > 0.04$, which is attributed to the influence of excess BKH contents, consistent with the study reported by Chen et al. [19]. It is well established that the phase structure significantly influences the electrical properties of materials. To systematically examine the changes in phase structure, the XRD pattern of the $(1-x)\text{KNLNS}-x\text{BKH}$ phase structure with varying x contents ($x = 0.0, 0.01, 0.02, 0.03, 0.04$, and 0.05) within the 2θ range of $44\text{--}47^\circ$ was fitted using the Lorentz function to represent the mixture of characteristic peaks of the O , R , or T phases. As displayed in Fig. 1(b), the KNLNS–BKH ceramics with $x = 0$ exhibit an orthorhombic structure with the splitting between the two diffraction peaks of $(022)_O$ and $(200)_O$ at 45.2° and 45.8° , respectively. As shown in Fig. 1(c), upon the addition of BKH at $x = 0.01$, the two split peaks gradually transform into $(022)_O$ at 45.2° (with low

relative intensity) and the $(200)_T$ peak (with high relative intensity), which are characteristic peaks of the tetragonal phase. In particular, the coexistence of orthorhombic and tetragonal phases was noted at $x = 0.01$. As illustrated in Fig. 1(d–e), the orthorhombic phase disappears as the BKH content increases from $x = 0.02$ to 0.04 , while the tetragonal phase becomes dominant. Notably, the rhombohedral phase appeared at $x = 0.04\text{--}0.05$ with a further increase in the BKH content, representing the characteristic $(200)_R$ peaks (Fig. 1(f–g)). Thus, the coexistence of tetragonal and rhombohedral phases occurred at $x = 0.03\text{--}0.04$, consequently improving their piezoelectric properties. This result is consistent with the findings reported by Liu et al. [18].

To further elucidate the phase transition, the XRD patterns of $(1-x)\text{KNLNS}-x\text{BKH}$ ceramics were analyzed using Rietveld refinement (Fig. 1(h)), with the corresponding refinement parameters summarized in Table 1. Rietveld refinements of $(1-x)\text{KNLNS}-x\text{BKH}$ ceramics were performed for compositions with $x = 0.00, 0.01, 0.02, 0.03, 0.04$, and 0.05 using mixed-phase models containing the $Amm2$ (orthorhombic), $P4mm$ (tetragonal), and $R3m$ (Rhombohedral) space groups. The fitting results for peak shape and peak position, structure, and background calculated in

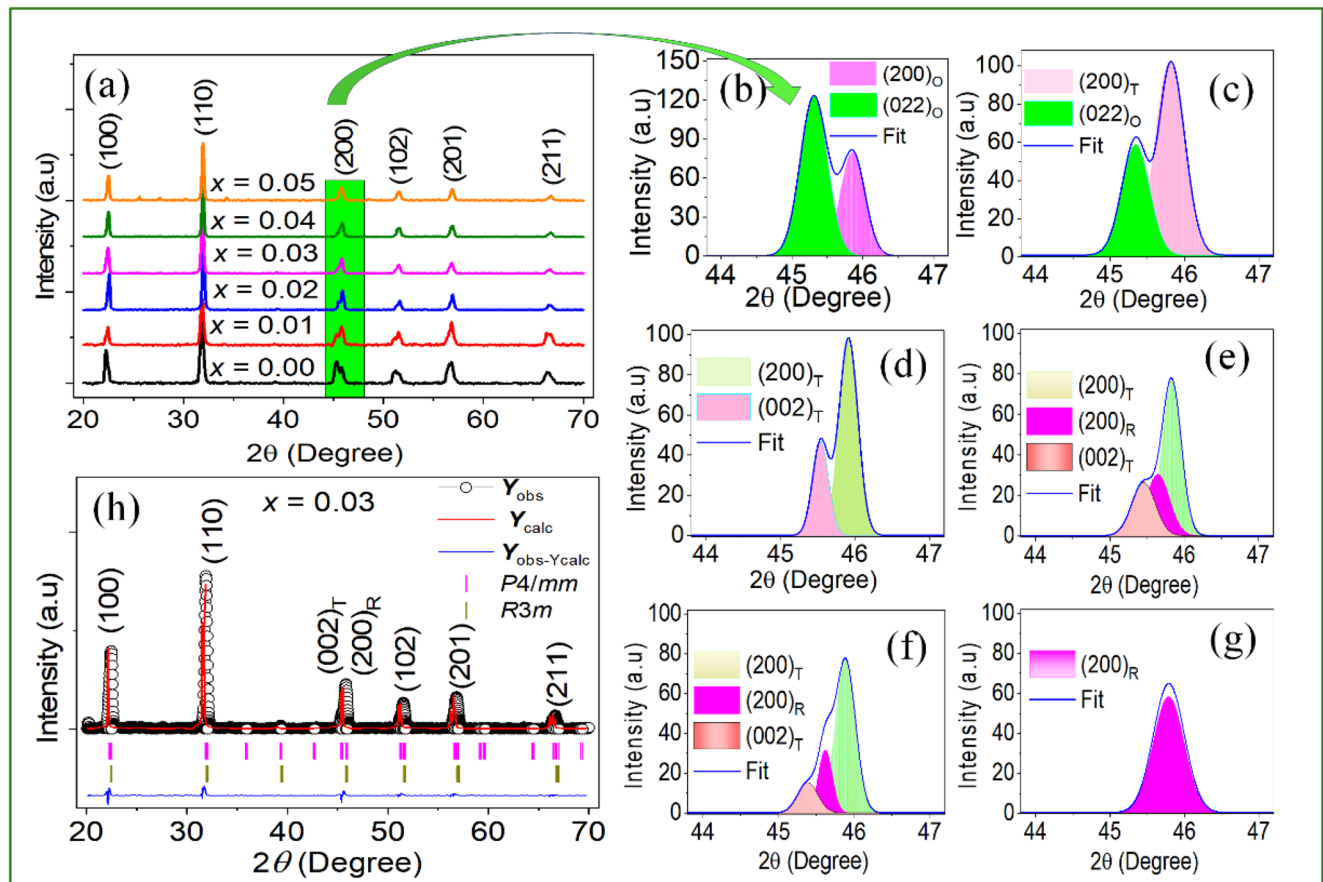
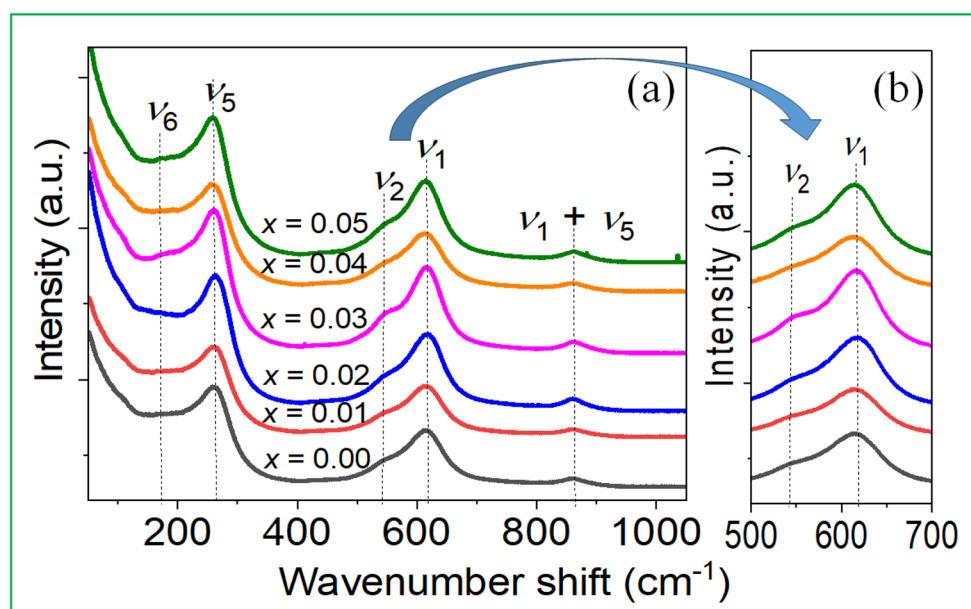


Fig. 1 XRD patterns of the $(1-x)\text{KNLNS}-x\text{BKH}$ ceramics in the 2θ range of (a) $20\text{--}70^\circ$; (b–g) $44\text{--}47^\circ$: (b) $x = 0.0$, (c) $x = 0.01$, (d) $x = 0.03$, (e) $x = 0.04$, (f) $x = 0.04$, (g) $x = 0.05$; (h) Rietveld-refined XRD patterns of samples at $x = 0.03$

Table 1 Lattice parameters and refinement parameters of the ((1-x)KNNs-xBKH ceramics obtained from the XRD Rietveld refinement

Sample	Space group	a (Å)	b (Å)	c (Å)	V (Å ³)	Phase fraction (%)	R _{wp} (%)	R _p (%)
x=0.00	<i>Amm2</i>	3.9840	5.6467	4.0170	90.3750	100	8.79	7.49
x=0.01	<i>Amm2</i>	3.9827	5.6423	4.0140	90.2010	44.44	7.91	7.72
	<i>P4mm</i>	3.9946	3.9946	3.9920	63.7000	55.56		
x=0.02	<i>Amm2</i>	3.9813	5.6380	4.0107	90.0260	24.49	7.97	8.05
	<i>P4mm</i>	3.9895	3.9895	3.9948	63.5820	75.51		
x=0.03	<i>P4mm</i>	3.9830	3.9830	3.9860	63.2350	53.85	8.1	8.14
	<i>R3m</i>	3.9800	3.9800	4.0210	55.1610	46.15		
x=0.04	<i>P4mm</i>	3.9779	3.9779	3.9903	63.1410	37.14	8.2	8.18
	<i>R3m</i>	3.9760	3.9760	4.0180	55.0090	62.86		
x=0.05	<i>R3m</i>	3.9720	3.9720	4.0150	54.8570	100	8.16	8.12

Fig. 2 Raman spectra of (1-x) KNLNS-xBKH ceramics with different x

terms of profile R_{wp} and R_p factors in Table 1 confirm the changes in phase structure with x increasing to form a new phase boundary. In particular, a rhombohedral-tetragonal phase ($R3m+P4mm$) is observed at $x=0.03$ – 0.04 which is expected to enhance the piezoelectric properties, consistent with the results reported by Huan et al. [20].

From a crystallographic perspective, the structural change to form a new phase boundary is due to the change in a , b , and c lattice parameters caused by the co-doping of Bi^{3+} and Hf^{4+} ions in $(\text{K}, \text{Na}, \text{Li})(\text{Nb}, \text{Sb})\text{O}_3$ ceramics. This is attributed to the competition between the larger radius of octahedrally coordinated B-site cations (Hf^{4+} , 0.72 Å) and the smaller A-site cation (Bi^{3+} , 1.03 Å) in $\text{Bi}_{0.5}\text{K}_{0.5}\text{HfO}_3$ compared with $(\text{K}_{0.48}\text{Na}_{0.48}\text{Li}_{0.04})(\text{Nb}_{0.95}\text{Sb}_{0.05})\text{O}_3$. Here, the cation radii are as follows: (K^+ , 1.38 Å; Li^+ , 0.92 Å; Na^+ , 1.32 Å; Nb^{5+} , 0.64 Å; Sb^{5+} , 0.62 Å). In other words, the structural transformation of the O -phase to the T -phase leads to changes in the lattice parameters of the ceramic when the $\text{Bi}_{0.5}\text{K}_{0.5}\text{HfO}_3$ impurity content is below 0.02 mol ($0.0 \leq x < 0.02$). Notably, an increase in x ($0.02 \leq x \leq 0.05$)

increases the lattice parameter c , thus promoting the transformation of the tetragonal phase (T -phase) to the rhombohedral (200_R) phase [21].

Figure 2 presents the Raman spectra of (1-x)KNLNS-xBKH ceramics measured at room temperature, illustrating the relationship between phase evolution and the role of BKH addition. Notably, all ceramic samples display three distinct scattering peaks in the studied frequency range of 50–1000 cm^{-1} . The major Raman shift peaks were located at around 190 cm^{-1} , 260 cm^{-1} , 580 cm^{-1} , 610 cm^{-1} , and 860 cm^{-1} . These peaks were well fitted to the vibration modes of $F_{2u}(\nu_6)$, $F_{2g}(\nu_5)$, $1E_g(\nu_2)$, $A_{1g}(\nu_1)$, and $F_{2g}(\nu_1 + \nu_5)$, respectively, indicating the characteristic perovskite structure of the KNN-based ceramics (Fig. 3). As reported in previous studies, the Raman shift peaks were located in the region below 200 cm^{-1} , which is associated with the A-site vibrations in the ABO_3 (Fig. 2) perovskite structure of the K^+ , Na^+ , Li^+ , and Bi^{3+} cations and rotational modes of the BO_6 octahedra [21–23]. Dwivedi et al. [23] suggested that the weak bands at 192 cm^{-1} in KNN ceramics are assigned

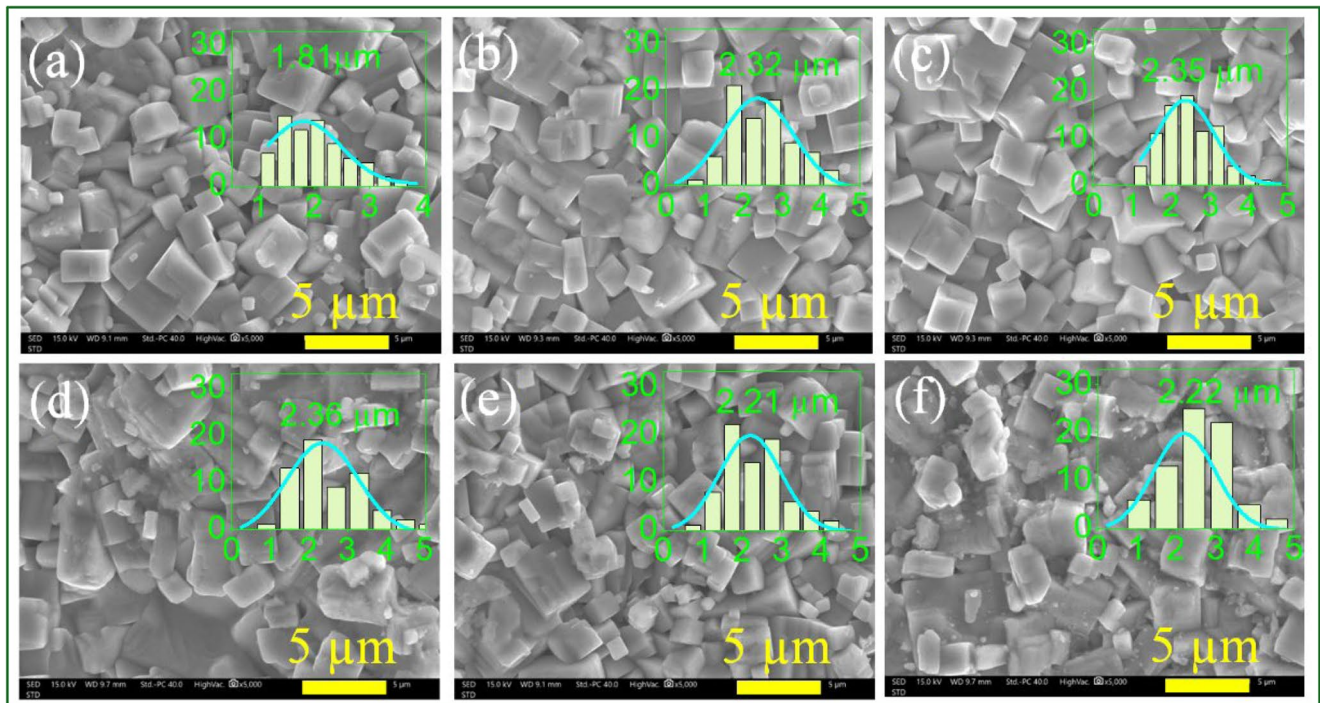


Fig. 3 Microstructure of (1-x)KNLNS-xBKH ceramics with varying x

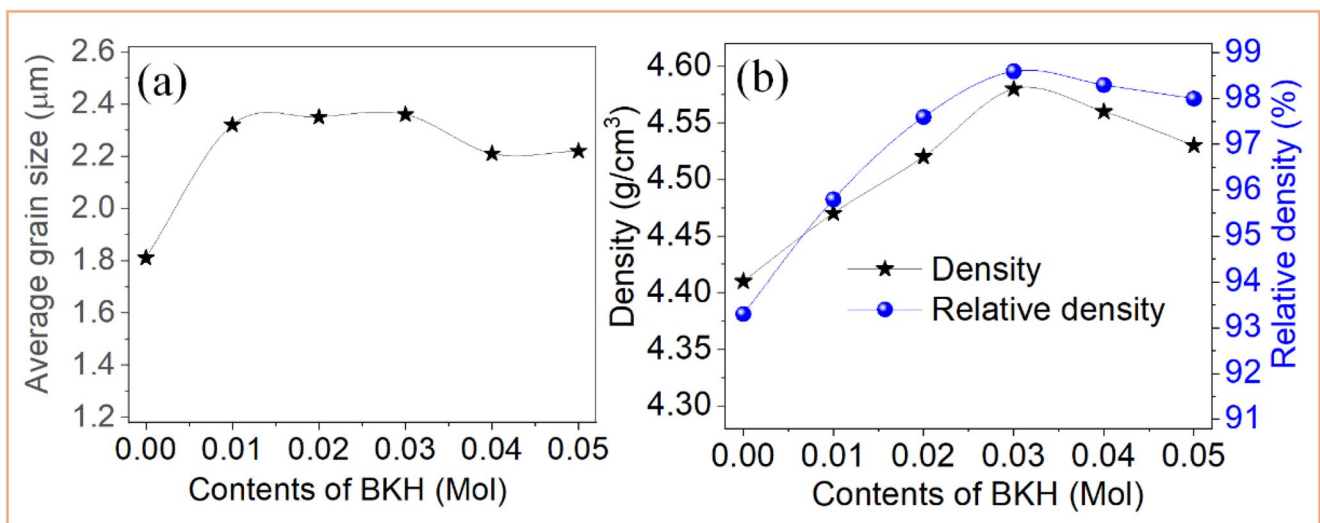


Fig. 4 Average grain size (a) and density (b) of (1-x)KNLNS-xBKH ceramics with varying x

to the translational modes of K^+ , Na^+ cations relative to NbO_6 octahedra. Moreover, as observed in Fig. 4, the peak positions of the ν_1 and ν_2 modes are the most prominent modes with relatively strong intensity, representing doubly degenerate symmetric O–Nb–O stretching vibrations. These peaks were fitted using a Gaussian function that is sensitive to slight changes in phase transition. Meanwhile, the ν_5 band originates from the triply degenerate symmetric O–Nb–O bending vibration of the NbO_6 octahedra in KNN-based ceramics. Moreover, the peaks tend to shift toward a

lower wave number with increasing BKH content in (1-x)KNLNS-xBKH ceramics; this observation is similar to that reported by Chen et al. [19]. This phenomenon is attributed to the weak distortion of the NbO_6 octahedron, resulting in higher structural symmetry; this finding is consistent with the XRD results and agrees with the report by Chen et al. [19]. As is well known, the A and B sites in $(K_{0.48}Na_{0.48}Li_{0.04})(Nb_{0.95}Sb_{0.05})O_3$ ceramics are occupied by K^+ (1.38 Å), Li^+ (0.92 Å), and Na^+ (1.32 Å) at the A site and Nb^{5+} (0.64 Å) and Sb^{5+} (0.62 Å) at the B site, respectively. The doping

of $\text{Bi}_{0.5}\text{K}_{0.5}\text{HfO}_3$ into $(\text{K}_{0.48}\text{Na}_{0.48}\text{Li}_{0.04})(\text{Nb}_{0.95}\text{Sb}_{0.05})\text{O}_3$ ceramics with ions of different radii (Bi^{3+} , 1.03 Å; and Hf^{4+} , 0.72 Å) causes structural distortion. Furthermore, the different molecular masses lead to a peak shift toward a lower wave number, resulting in a corresponding variation in the crystal cell volume, as observed in the XRD pattern of KNLNS–BKH ceramics. Notably, similar Raman spectral behavior has been observed in other KNN-based ceramics, further confirming the existence of a change in the phase structure symmetry [23].

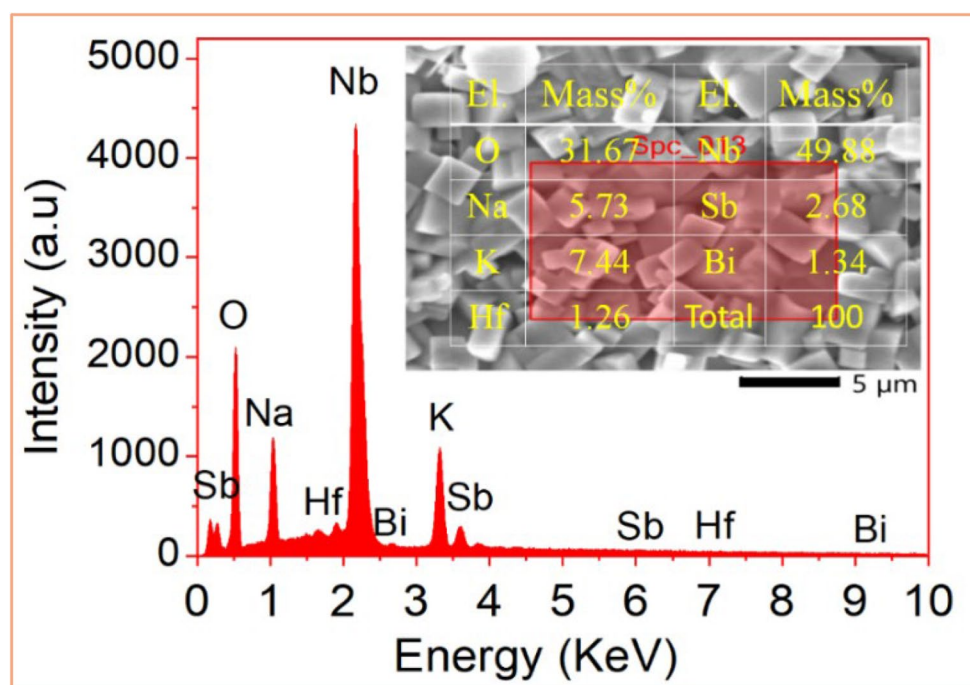
Figure 3 displays the SEM images of $(1-x)\text{KNLNS}-x\text{BKH}$ ceramics, with the grain distribution figures shown in the insets for $x=0.0, 0.01, 0.02, 0.03, 0.04$, and 0.05 . As observed in Fig. 4, the grains in all samples exhibit a well-developed microstructure with clearly defined grain boundaries. The micrographs reveal that the grain size increases with increasing x content and reaches a maximum of approximately $2.36\text{ }\mu\text{m}$ at $x=0.03$ (Fig. 4(d)), and subsequently decreases with further increases in x (Fig. 4(a)). The phenomenon is attributed to the solubility limit of $(\text{Bi}_{0.5}\text{Na}_{0.5})\text{HfO}_3$ into the KNLNS ceramics, similar to the findings reported by He et al. [24]. This can be explained by the dissolution of Bi and Hf present in $(\text{Bi}_{0.5}\text{Na}_{0.5})\text{HfO}_3$ into the KNLNS crystal lattice. The Hf^{4+} ions (0.72 Å) enter the KNLNS crystal lattice and substitute for Nb^{5+} (0.64 Å) and Sb^{5+} (0.62 Å) ions, while the Bi^{3+} ions (1.03 Å) substitute for K^+ (1.38 Å), Li^+ (0.92 Å), or Na^+ (1.32 Å) ions due to their similar ionic radii. Consequently, the integration of BKH into the KNLNS ceramics promotes grain growth, facilitates the filling of small grains between large grains, enhances the ceramic density (Fig. 4(b)) and further bolsters the piezoelectric properties. However, the grain size gradually decreases when the BKH doping contents exceed 0.03 mol. The grains become highly refined when $x>0.04$, which is attributed to the high content of BKH aggregates at the grain boundary that inhibit grain growth. Moreover, pores appear in the sample when $x>0.03$, indicating a deterioration in ceramic density; this finding agrees with the ceramic density measurement results (Fig. 4(b)). As shown in Fig. 4(b), the density of $(1-x)\text{KNLNS}-x\text{BKH}$ ceramics varies with the x content sintered at $T_2 = 1050\text{ }^\circ\text{C}$ for 4 h. It is observed that the average ceramic density increases with increasing x content, reaching a maximum value of 4.58 g/cm^3 at $x=0.03$, and then decreases. The increase in ceramic density is attributed to enhanced crystallization and the formation of uniformly distributed and closely packed grains. The densification mechanism results from thermal shrinkage, which is related to the microstructure of the samples. This implies that the incorporation of BKH into the KNLNS ceramics improves the microstructure of ceramics by filling small grains between large grains, thereby enhancing the ceramic density as discussed above. Typically, the ceramic density

of the KNN-based ceramics ranges from 4.3 to 4.6 g/cm^3 , which corresponds to a relative density of 93.3–98.1% of the theoretical value [4, 22, 23]. According to previous studies on KNN-based ceramics, the grain shape of the ceramics is square, suggesting that the pores cannot be eliminated [18, 19, 24]. Compared with these studies, our KNLNS–BKH ceramics exhibit a significantly improved microstructure. In other words, the well-developed microstructure with clearly defined grain boundaries results in small grains that fill the void between large grains, consequently increasing the density and improving the electrical properties [24].

Figure 5 presents the EDS spectrum of 0.98KNLNS–0.02BKH ceramics sintered at $1050\text{ }^\circ\text{C}$ for 4 h. As depicted in Fig. 6, the homogeneous distribution of all elements, including Na, K, Nb, Sb, Bi, Hf, and O, is consistent with the theoretical design and confirms the successful diffusion of Bi and Hf elements into the KNLNS lattice. Additionally, as shown in Fig. 7(a), the elements were evenly distributed, overlapping the surface of the 0.98KNLNS–0.02BKH ceramic sample. Moreover, it is demonstrated that the high-intensity area of the Sb maps covered the low-intensity area of the Nb map, whereas the high-intensity area of the Bi covered the low-intensity area of the Hf. As illustrated in Fig. 7(b–h), the high-intensity areas of the K, Na, and Bi maps corresponded to those of the Nb, Sb, and Hf, which filled the pores and created a densely packed microstructure of 0.98KNLNS–0.02BKH ceramic. This is consistent with studies for KNN-based materials [4, 25].

Fig. 7 illustrates the dependence of the dielectric constant and dielectric loss of $(1-x)\text{KNLNS}-x\text{BKH}$ ceramics with different x , measured at 10 kHz at room temperature. Fig 7 shows that the dielectric constant (ϵ_r) gradually increases with increasing BKH content and reaches a maximum value of $\epsilon_r=1198$ at $x=0.03$ mol. In contrast, the dielectric loss ($\tan\delta$) decreases to a minimum value of $\tan\delta=0.04$ at the same composition of $x=0.03$ mol. A further increase in the x content leads to a decrease in ϵ_r and an increase in $\tan\delta$. These results indicate that the sample containing 0.03 mol BKH exhibits superior dielectric properties compared to the other samples. According to Ullah et al. [13], the enhancement of dielectric properties is attributed to higher ceramic density, larger grain size, and improved microstructure. He et al. [24] suggested that increasing grain size promotes the switching of domains, resulting in enhanced electrical properties. In our opinion, the change in phase structure from orthorhombic to tetragonal to rhombohedral phases produces different types of domains. Consequently, electrical properties exhibit variation trends under the influence of domains. In general, the improvement in the electrical properties of ceramics is attributed to the grain size and phase boundary effects [22]. Thus, the values of ϵ_r and $\tan\delta$ can be well enhanced when the BKH complex content is

Fig. 5 EDS analysis of 0.98) KNLNS-0.02BKH ceramic sample



incorporated into KNLNS ceramics. It is noted that the electrical properties of KNLNS-based ceramics are highly sensitive to preparation conditions, similar to the report of Tao et al. [16].

As shown in Fig. 8(a), the dielectric constant and dielectric loss of (1-*x*)KNLNS-*x*BKH ceramics at varying temperatures (25–400 °C) were measured at 10 kHz to observe the dependence of dielectric properties on temperature. Furthermore, the (ϵ -*T*) curves of the (1-*x*)KNLNS-*x*BKH ceramics in the range of *x*=0–0.05 exhibit two clear peaks above room temperature: the first one at low temperatures, corresponding to the *O*-*T* phase transition temperature (T_{O-T}), and the second one at higher temperatures, corresponding to the *T* phase to cubic phase transition temperature (T_C). Figure 8(b) demonstrates this trend on the dielectric loss curve. It is noteworthy that when the value of *x* increases to 0.04, the ceramics continue to maintain a relatively high T_C (>300 °C), which can be applied in high-temperature environments. As depicted in Fig. 8(a), both T_{O-T} and T_C shifted towards the lower temperature side, and a broadening of these transitions is observed with increasing BKH content, which is attributed to the difference in the ionic radius of Bi³⁺ and Hf⁴⁺ ions with the host ions. The ionic radius of Bi³⁺ (1.03 Å) is comparable to those of K⁺ (1.38 Å), Li⁺ (0.92 Å), or Na⁺ (1.32 Å) ions, whereas that of Hf⁴⁺ (0.72 Å) ions is similar to those of Nb⁵⁺ (0.64 Å) and Sb⁵⁺ (0.62 Å) ions. It is believed that the substitution of the [Bi_{0.5}K_{0.5}]²⁺ species decreased T_{O-T} , whereas Hf⁴⁺ ions increased the T_{R-O} corresponding to the KNLNS-BKH ceramics, so the coexistence of *O*-*T*, *R*-*O*-*T*, and *R*-*T* phases was observed near room temperature, similar to the report of Liu et al.

[18]. According to the research results of Liu et al. [18] for (Bi_{0.5}Na_{0.5})HfO₃-doped (K_{0.4}Na_{0.6})(Nb_{0.96}Sb_{0.04})O₃ ceramics, the T_C and T_{O-T} decrease with increasing BKH content, whereas the T_{R-O} rises, resulting in T_{O-T} and T_{R-O} achieving a merger at the BKH content of 0.035 mol. This behavior stems from the high temperature-sensitive nature of pure KNN ceramics in the range of -150 to 420 °C, including rhombohedral-orthorhombic phase transition (T_{R-O} ~125 °C), orthorhombic-tetragonal phase transition (T_{O-T} ~200 °C), and tetragonal-cubic phase transition at (T_C ~420 °C) [18, 26].

In the present study, the BKH dopant is effective in reducing the T_{O-T} temperature to near room temperature. As reported by Liu et al., the T_{O-T} values of the (1-*x*)KNLNS-*x*BKH (*x*=0.0, 0.01, 0.02, 0.03, 0.04, and 0.05) ceramics were in the range of 70–40 °C (Fig. 8(c)) [18]. As shown in Fig. 8(c), the $\epsilon_{\max}$ values at T_C increased with the BKH content, reaching a maximum value of 6328 for *x*=0.03, followed by a subsequent decrease. These results indicate that the dielectric properties of the sample containing 0.03 mol BKH are superior to those of the other samples. This is attributed to the high ceramic density and the larger grain size, which facilitate easier domain orientation and lead to an increased $\epsilon_{\max}$ value. Furthermore, the change in phase structure from orthorhombic to tetragonal to rhombohedral phases produces different types of domains, resulting in improved dielectric properties of ceramics as discussed above. However, as presented in Figs. 7 and 8, the dielectric properties tend to decrease when the BKH content increases beyond 0.03 mol. This phenomenon arises from the solubility limit of Bi³⁺ and Hf⁴⁺ ions within the KNLNS matrix.

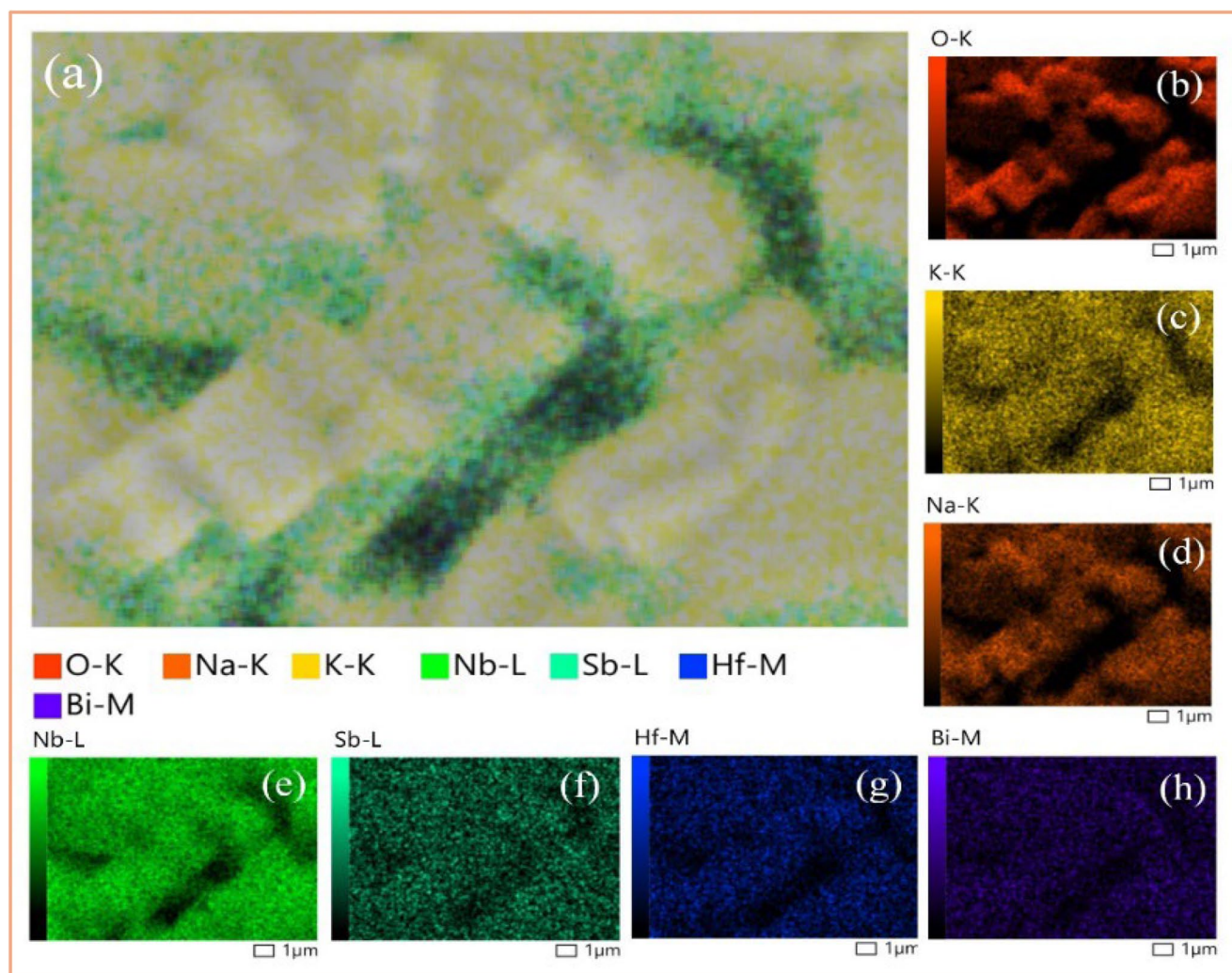


Fig. 6 Element mappings of 0.98KNLNS-0.02BKH ceramic sample: O, K, Na, Nb, Sb, Hf and Bi

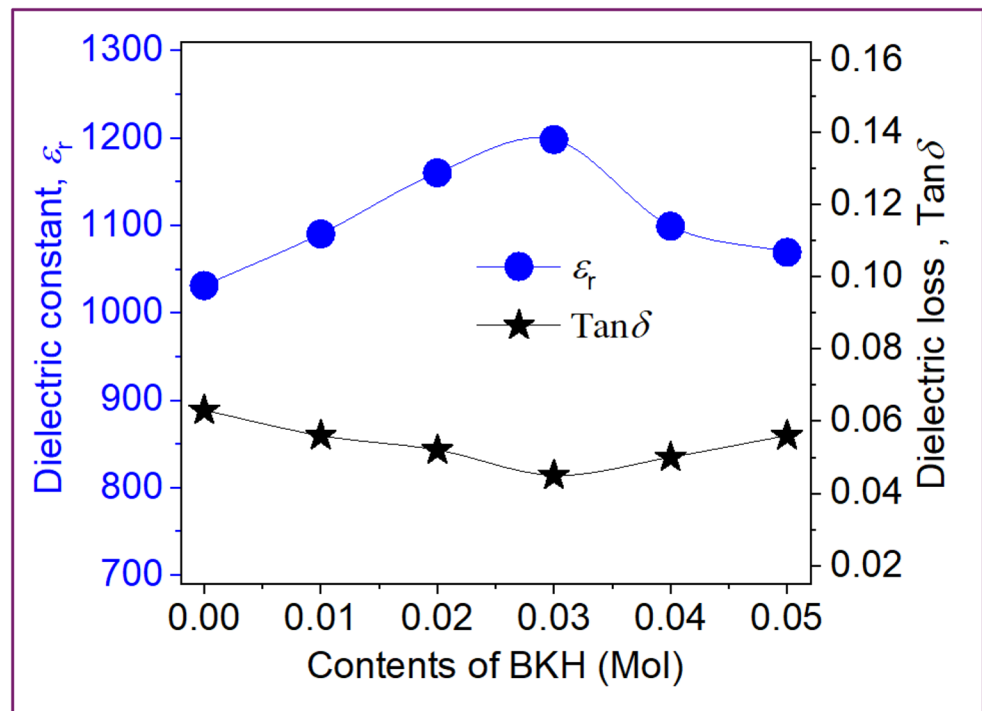
These results can be attributed to the effect of the R - T phase boundary when the BKH content increases beyond 0.03 mol. Besides, the decrease in ceramic grain size and irregular microstructure at high BKH contents is also responsible for the reduction in electrical properties of the materials.

To examine the influence of BKH contents on relaxor behavior in the $(1-x)\text{KNLNS}-x\text{BKH}$ material, the diffusivity values (γ) were evaluated by plotting $\ln(1/\varepsilon - 1/\varepsilon_{\max})$ versus $\ln(T-T_m)$ at 10 kHz, as presented in Fig. 8(d). Notably, these γ values vary with different x content for ceramic samples. The γ value increases from 1.38 to 1.53 as the BKH contents rise from x to 0.04 and subsequently decreases to 1.48 with further increases up to $x=0.05$. This is due to the larger ionic radius of Hf^{4+} (0.72 Å) compared with Nb^{5+} (0.64 Å) and Sb^{5+} (0.62 Å), which increases the disorder in the lattice structure. According to Li et al. [21], the diffuse phase transition behavior is attributed to increased lattice distortion and the formation of polar nanometer-sized regions caused by $\text{Bi}_{0.5}\text{K}_{0.5}\text{HfO}_3$. These features are beneficial for enhancing

their piezoelectric properties and will be discussed in the next section. However, Fig. 8(d) shows the value of γ in the range of 1.38–1.53, suggesting a comparatively low degree of relaxation. This indicates that the addition of BKH does not cause serious phase segregation within the studied composition in the range of 0–0.05 mol, consistent with the findings of reference [18].

Figure 9(a–c) presents polarization versus temperature, under different electric fields for d_{33} and k_p values of the 0.98KNLNS–0.02BKH sample to optimally exploit the role of polarization conditions. As shown in Fig. 9(a–c), the 0.98KNLNS-0.02BKH ceramic exhibited the best piezoelectric properties with $d_{33}=232$ pC/N and $k_p=0.39$ at the polarization temperature of 45 °C, in accordance with the O - T phase transition temperature. This result indicates that the 0.98KNLNS-0.02BKH ceramic exhibits polymorphic phase transition behavior [8, 27], with the coexistence of the T and O phases at ~ 45 °C, which enhances the piezoelectric properties, in accordance with

Fig. 7 Dielectric constant and dielectric loss of (1-*x*)KNLNS-*x*BKH ceramics with varying *x*.



the results of the dielectric properties study mentioned above. Based on these observations, the polarization conditions of the (1-*x*)KNLNS-*x*BKH ceramic samples were selected as 35 kV/cm at 45 °C for 25 min. The piezoelectric parameters, including k_p , k_t , and d_{33} , were measured as a function of composition *x*, as shown in Fig. 9(d). It is noted that k_p , k_t , and d_{33} exhibit an increasing trend with increasing BKH content, reaching their respective maximum values at $x=0.03$ with $k_p = 0.41$, $k_t = 0.45$, and $d_{33} = 238$ pC/N. The significant improvement in the piezoelectric properties of ceramics is attributed to the formation of new phase boundaries, as discussed above. At $x=0.03$, the composition is situated near the *R-T* phase boundary, which provides more possible directions of spontaneous polarization of the domains, facilitating easier polarization, as reported by Liu et al. [18]. These authors suggest that compared with other phase boundaries, such as the *R-O* phase boundary, the *R-T* phase boundary generally exerts a stronger enhancing effect on the piezoelectric responses [18]. We propose that the improvement in piezoelectric properties results from microstructural refinement with large grain size, small grain boundaries, high ceramic density, and phase boundary effects. In other words, the increased grain size promotes the switching of domains, resulting in enhanced piezoelectric properties, similar to those reported by He et al. [24]. Hence, the piezoelectric properties of (1-*x*)KNLNS-*x*BKH reach their maximum at $x=0.03$, corresponding to the *R-T* phase boundary, where the highest density, the most uniform grain size, and the tightest grain bonding facilitate easier

domain reorientation and significantly promote domain wall motion.

As plotted in Fig. 10(a), the ferroelectric properties of the (1-*x*)KNLNS-*x*BKH ceramics with varying *x* were determined from their ferroelectric hysteresis loops measured using the Sawyer-Tower method. Notably, a typical and well-saturated ferroelectric hysteresis loop was observed at a low electric field of nearly 20 kV/cm for the (1-*x*)KNLNS-*x*BKH ceramics with $x=0.0$, 0.01, 0.02, 0.03, 0.04, and 0.05. Furthermore, the squareness of the hysteresis loop (R_{sq}) of (1-*x*)KNLNS-*x*BKH ceramics with $x=0.0$, 0.01, 0.02, 0.03, 0.04, and 0.05 initially increased slightly from 0.91 to 0.95 as *x* increased from 0 to 0.03 and then decreased sharply as *x* continued to increase (Fig. 10(b)). The R_{sq} values were 1.90, 1.91, 1.92, 1.95, 1.46, and 1.28 for BKH contents of 0.0, 0.01, 0.02, 0.03, 0.04, and 0.05 mol, respectively, which were consistent with the results of the diffusion phase transition behavior of materials discussed earlier. As shown in Fig. 10(b), the remanent polarization (P_r) and coercive field (E_c) were determined from the hysteresis loops of the samples.

Figure 10(b) shows the dependence of P_r and E_c on *x* content in (1-*x*)KNLNS-*x*BKH ceramics. Notably, the P_r value displays composition dependence. It initially rises and reaches maximum values at $x=0.03$, a composition near the *R-T* phase boundary, with $P_r = 15.4$ $\mu\text{C}/\text{cm}^2$, then decreases with further increases in the BKH content. Meanwhile, the coercive field (E_c) of the samples tends to decrease with increasing *x*. The increase in the values of P_r is attributed to the rise in BKH content, leading to a change in the crystal

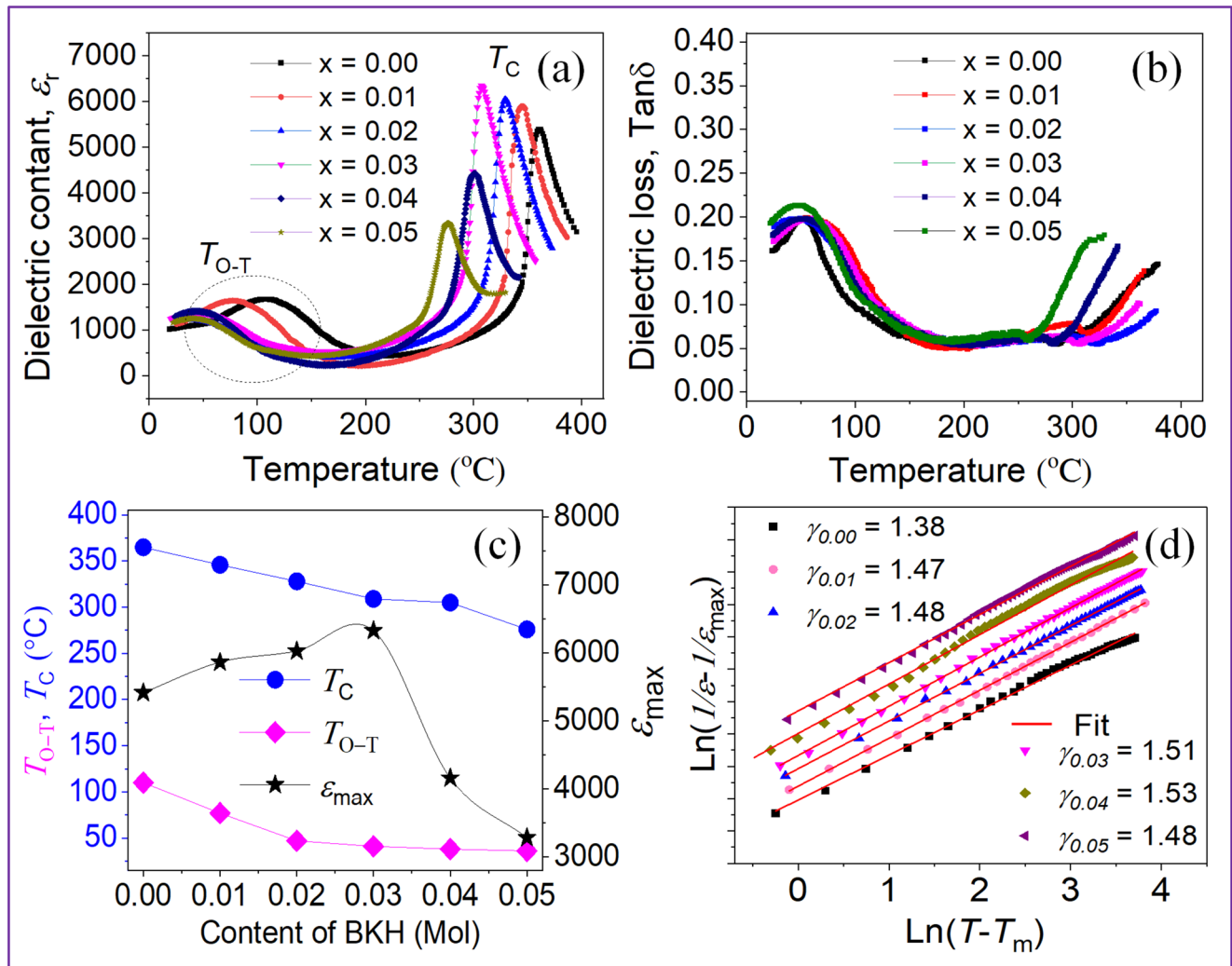


Fig. 8 The dependence of dielectric properties on temperature of (1-x)KNLNS-xBKH ceramics

structure to form the new phase boundary in the ceramics, and consequently enhances the likelihood of polarization. It is well known that increasing the substitution of Bi^{3+} and Hf^{4+} ions into (K, Na)(Nb, Sb) O_3 leads to a continuous decrease in T_C and T_{O-T} , whereas the T_{R-O} gradually increases to room temperature. This results in a gradual shrinkage of the orthorhombic phase region, causing T_{O-T} and T_{R-O} to merge at $x=0.03$. Consequently, this forms an R - T phase boundary that improves their ferroelectric properties, consistent with the report of Liu et al. [18]. However, further increasing the BKH content to 0.04 mol led to a reduction in the ferroelectric properties of the ceramics, which is attributed to Bi^{3+} and Hf^{4+} ions exceeding the solubility limit in the (K, Na)(Nb, Sb) O_3 matrix. This can be explained by the fact that the coexistence of the R , O , and T phases leads to a lower energy barrier in the ceramics at the phase boundary region, facilitating easier polarization rotation. This enhances their ferroelectric properties, resulting in a high P_r and low E_c .

Nevertheless, these values decrease as the (1-x)KNLNS-xBKH component moves away from the phase boundary.

Recently, energy storage properties have received considerable research attention, particularly in thin-film ferroelectric materials [7, 28]. In this study, we calculated the parameters related to the energy storage properties of bulk (1-x)KNLNS-xBKH ($x=0.0, 0.01, 0.02, 0.03, 0.04$, and 0.05) ceramic materials, including the recoverable energy storage density (W_{rec} , the model in the inset of Fig. 11(a)) and the energy storage efficiency (η), using Eqs. 1 and 2, respectively.

$$W_{\text{rec}} = \int_{P_r}^{P_{\text{max}}} E dP \quad (1)$$

$$\eta = \frac{W_{\text{rec}}}{W_{\text{rec}} + W_{\text{loss}}} \quad (2)$$

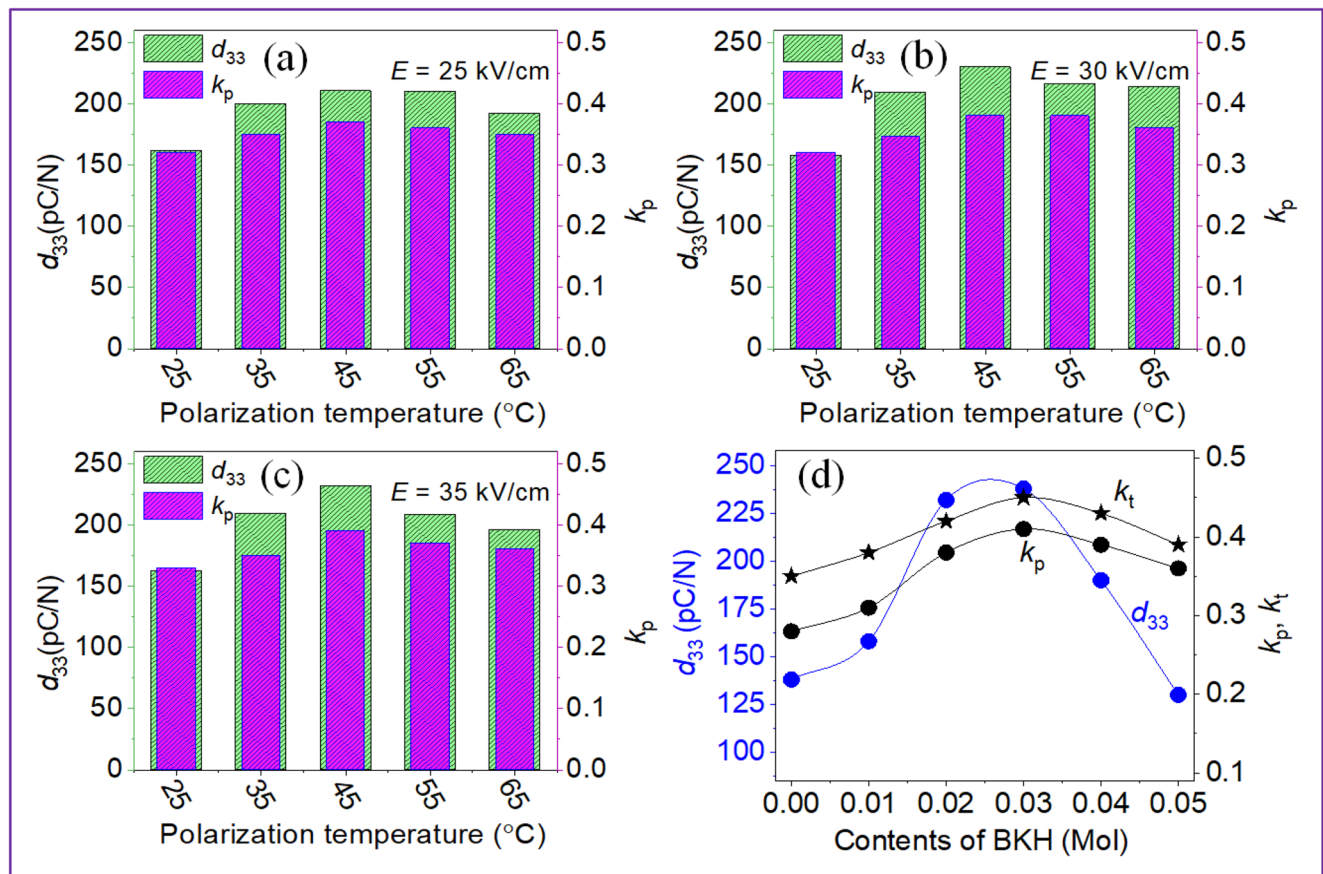


Fig. 9 (a–c) Polarization versus temperature, under different electric fields for d_{33} and k_p values of the 0.98KILNS–0.02BKH sample; (d) The piezoelectric parameters (k_p , k_t , and d_{33}) of the $(1-x)\text{KNLNS}-x\text{BKH}$ ceramic with varying x

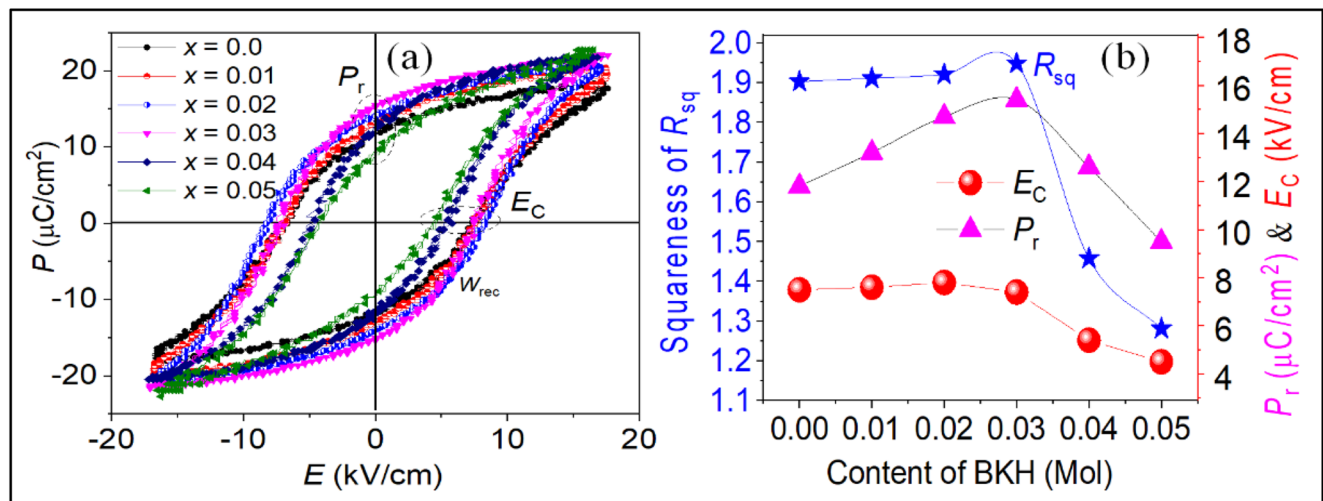


Fig. 10 The P - E hysteresis loops of the $(1-x)\text{KNLNS}-x\text{BKH}$ ceramic with varying x

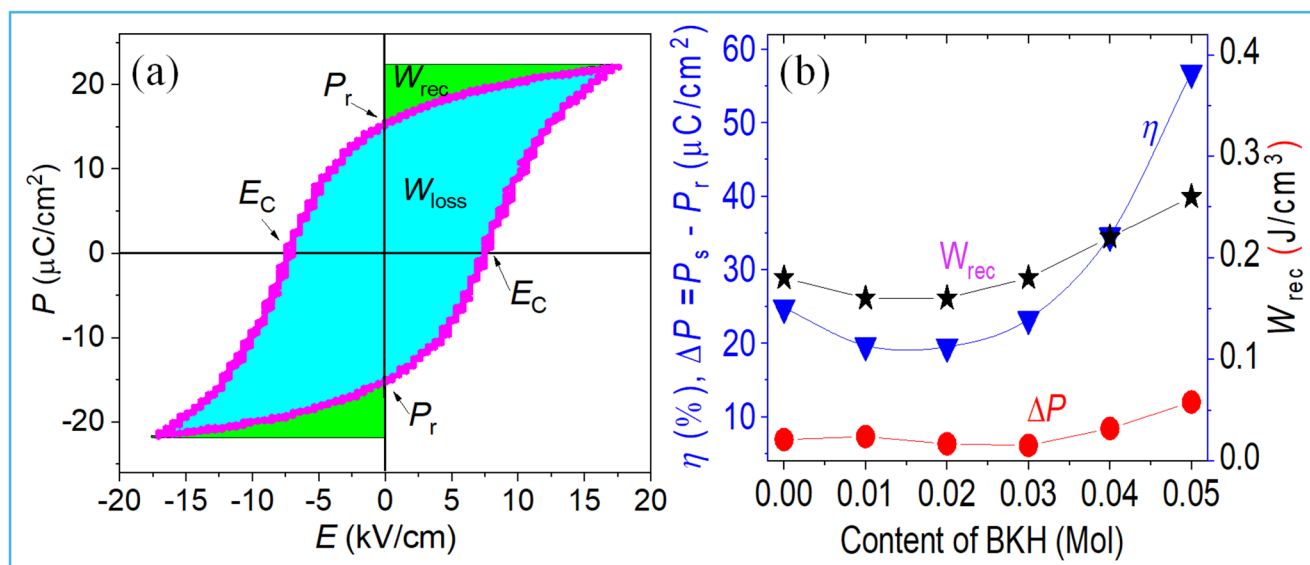


Fig. 11 (a) Schematic diagram; (b) Calculated W_{rec} and η values of the energy storage properties of $(1-x)\text{KNLNS}-x\text{BKH}$ ceramic with different x

The results are plotted in Fig. 11(b). As observed in Fig. 11(b), both W_{rec} and η decreased slightly as the BKH content increased from 0 to 0.03 mol, and then increased significantly, reaching maximum values of 0.26 J/cm^3 and 56.8%, respectively, at $x=0.05$. As shown in Fig. 11(b), the improvement in energy storage density depends on the difference between the high polarization saturation (P_s) and low remnant polarization (P_r) ($\Delta P = P_s - P_r$). In other words, P_r increases when the BKH content is between 0.0 and 0.03 mol, leading to a decrease in ΔP , thereby reducing W_{rec} and η . Nevertheless, ΔP increases when x increases to 0.04 and 0.05, leading to an increase in the W_{rec} and η values, as shown in Fig. 11(b). Additionally, the improvement in energy storage properties is attributed to the enhancement of ceramic relaxation properties, as suggested by previous studies [4, 16], which is consistent with the results of the diffusivity values (γ) shown in Fig. 8(d). Overall, a high energy storage efficiency (η) of 56.8% is achieved for KNLNS–BKH ceramics at $x=0.05$ under a low electric field of 20 kV/cm , broadening their potential applications, including dielectric, ferroelectric, piezoelectric, and energy storage properties.

4 Conclusion

In conclusion, the $(1-x)(\text{K}_{0.48}\text{Na}_{0.48}\text{Li}_{0.04})(\text{Nb}_{0.95}\text{Sb}_{0.05})\text{O}_3 - x\text{Bi}_{0.5}\text{K}_{0.5}\text{HfO}_3$ ($x=0.0, 0.01, 0.02, 0.03, 0.04$, and 0.05) ternary ceramics were examined in this study by varying the $\text{Bi}_{0.5}\text{K}_{0.5}\text{HfO}_3$ component to develop a novel composition material using a two-step sintering method. Herein, the phase boundaries (O - T and R - T) were successfully

formed by adjusting the $\text{Bi}_{0.5}\text{K}_{0.5}\text{HfO}_3$ content in the range of 0–0.05 mol, and the enhanced electrical properties were subsequently achieved at the optimal composition. At $x=0.03$, the 0.97KNLNS–0.03BKH ceramics exhibited good electrical properties (density = 4.58 g/cm^3 ; $\epsilon_r = 1198$; $\epsilon_{\text{max}} = 6328$; $k_p = 0.40$; $k_t = 0.45$; $d_{33} = 238$ pC/N ; and $P_r = 15.4$ $\mu\text{C}/\text{cm}^2$). Furthermore, the dielectric relaxation behavior of $(1-x)\text{KNLNS}-x\text{BKH}$ ceramics was investigated for different x contents, achieving a high Curie temperature ($T_C = 310$ $^\circ\text{C}$) and a low orthorhombic-tetragonal phase transition ($T_{O-T} = 40$ $^\circ\text{C}$) at $x=0.03$. Specifically, the ceramics with $x=0.05$ demonstrated a high recoverable energy density ($W_{\text{rec}} = 0.26$ J/cm^3) and a large energy storage efficiency ($\eta = 56.8\%$) under a low electric field of 20 kV/cm , demonstrating significant potential for the practical applications of KNLNS–BKH ceramics.

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Author contributions Experimental design, and writing the original manuscript by Vuong L. D., Cuong N. X., Nhat N. D., and Hieu L. D. were involved in planning and supervising the work. Kieu V. T. T. and Tan L. V. processed the experimental data. Data analysis, the investigation by Dat, T. N., Wei. P-C, and Tung V.T. Theoretical analysis, review and editing by Chuc N. H. T. and Nha V. Q. All authors discussed and approved the final manuscript.

Data availability The raw data supporting the conclusions of this article will be made available by the authors on reasonable request.

Declarations

Competing interests The authors declare no competing interests.

References

1. L.D. Vuong, N. Van Thinh, D.V. On, V.T. Tung, *J. Mater. Sci.: Mater. Electron.* **36**, 284 (2025)
2. W. Liu, T. Zheng, Z. Zhou, Y. Ding, Y. Qin, Z. Fu, X. Ruan, Z. Gao, X. Lv, J. Wu, *Nat. Commun.* **16**, 1527 (2025)
3. X. Zhou, H. Gong, J. Zhang, J. Liu, D. Zhang, Y. Zhang, *J. Mater. Chem. A* **13**, 40689–40697 (2025)
4. L.T.U. Tu, L.D. Vuong, T.N. Dat, V.T. Tung, *J. Mater. Sci.* **131**, 210 (2025)
5. P.D. Gio, L.D. Vuong, V. ThanhTung, *J. Electroceram.* **46**, 107–114 (2021)
6. N. Van Thinh, T. Van Chuong, T.N. Dat, V.T. Tung, *J. Ceramic Process. Res.* **24**, 478–485 (2023)
7. L.D. Vuong, N.Q. Lich, N.X. Cuong, V.Q. Nha, N.D. Nhat, L.D. Hieu, L.P. Son, H.T.T. Linh, N.H. Chuc, T.N.A. Tuyen, T.N. Dat, P.-C. Wei, *Mater. Res. Express.* **10**, 105701 (2023)
8. B. Li, C. Xiong, X. Cao, X. Wei, Y. Qiu, D. Gao, *Appl. Phys. Lett.* **126** (2025)
9. L. Bian, Q. Wang, S. He, J. Wu, K. Li, Y. Liu, B. Yang, *J. Eur. Ceram. Soc.* **44**, 5048–5054 (2024)
10. L.T.U. Tu, P.D. Gio, *J. Mater. Sci.: Mater. Electron.* **34**, 217 (2023)
11. L.T.U. Tu, N.T. Tho, *J. Mater. Sci.: Mater. Electron.* **34**, 917 (2023)
12. P.D. Gio, L.D. Vuong, L.T.U. Tu, *J. Mater. Sci.: Mater. Electron.* **32**, 13738–13747 (2021) <https://doi.org/10.1007/s10854-021-05951-1>
13. P.D. Gio, H.Q. Viet, L.D. Vuong, *Int. J. Mater. Res.* **109**, 1071–1076 (2018)
14. J. Zhang, X. Sun, W. Su, W. Yao, C. Zhou, *Scripta Mater.* **176**, 108–111 (2020)
15. P. Li, J. Zhai, B. Shen, S. Zhang, X. Li, F. Zhu, X. Zhang, *J. Mater. Sci.* **30**, 1705171 (2018)
16. H. Tao, W. Wu, J. Wu, *J. Alloys Compd.* **689**, 759–766 (2016)
17. Y. Chen, D. Xue, M. Shi, X. Jiang, Z. Chen, X. Liu, G. Liu, Z. Xu, *J. Am. Ceram. Soc.* **100**, 3920–3927 (2017)
18. Y. Liu, Y. Yi, Y. Yu, Y. Pan, C. He, X. Liu, Z. Xu, G. Liu, Y. Chen, *Ceram. Int.* **45**, 6328–6334 (2019)
19. X. Chen, J. Sun, X. Li, X. Yan, X. Li, J. Shi, C. Sun, F. Pang, H. Zhou, *J. Mater. Sci.: Mater. Electron.* **30**, 16407–16414 (2019)
20. Y. Huan, X. Wang, W. Yang, L. Hou, M. Zheng, T. Wei, X. Wang, *J. Adv. Ceram.* **11**, 935–944 (2022)
21. B. Li, C. Xiong, G. Gao, D. Gao, *J. Mater. Sci.: Mater. Electron.* **35**, 153 (2024)
22. L.T.U. Tu, L.D. Vuong, N.X. Cuong, N.Q. Lich, N.D. Nhat, L.D. Hieu, V.T.T. Kieu, T.N. Dat, P.-C. Wei, *J. Mater. Sci.: Mater. Electron.* **35**, 1363 (2024)
23. S. Dwivedi, T. Pareek, S. Kumar, *RSC Adv.* **8**, 24286–24296 (2018)
24. B. He, Y. Du, Y. Liu, J. Wang, W. Liu, H. Xu, *J. Mater. Sci.: Mater. Electron.* **32**, 18240–18250 (2021)
25. Y. Zhou, B. Fang, S. Zhang, X. Lu, J. Ding, *J. Korean Ceram. Soc.* **59**, 551–564 (2022)
26. H. Tao, C. Liao, Z. Liu, B. Wu, C. Zhao, W. Wu, L. Zhao, J. Ma, *J. Mater. Chem. C* **13**, 10242–10250 (2025)
27. N. Truong-Tho, D. Le Vuong, *J. Mater. Sci.: Mater. Electron.* **32**, 16601–16611 (2021)
28. R. Singh, R. Nandan, S. Pathak, S. Rao, A. Kumar, R. Sharma, S. Sinha, H.A. Laasri, R. Noorjahan, Nagarajan, *J. Alloys Compd.* **184522** (2025)

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